**1992 Progress Report for ONR grant No. N000014-91-J-1502: Defects and Disordered Solids** by Clare Yu, Physics Department, UC Irvine, Irvine, CA

Our research has concentrated on strongly interacting defects in solids in an effort to better understand glasses, random magnetic systems and defects in crystals. Most of my effort went into a Monte Carlo simulation of randomly placed defects with internal degrees of freedom which interact via elastic strain fields. This has been proposed as a model of the low temperature properties of glasses.[1] The defects were represented by stress tensors. To model the defect degrees of freedom, each tensor component was treated like an Ising spin, i.e., it could be positive or negative according to a Boltzmann weighting factor. The interactions were the tensor analog of a dipolar interaction, i.e., they went as $1/r^3$ and had angular dependence. Using finite size scaling, I found the interesting and unusual result that two spin glass phase transitions occur: one for the diagonal components of the defect stress tensor and the other for the off-diagonal components. In addition examination of the fourier transform of the quenched ground state revealed the intriguing result that the off-diagonal components have planar antiferroelastic correlations while the diagonal components do not. I predicted that the signature of these transitions is the divergence of the fourth order elastic susceptibilities at the transition temperatures. However, it will take more work to understand how these divergences affect the elastic constants that can be measured by ultrasound. A preprint of this work has been submitted to Physical Review Letters.

As I discussed in my proposal, a system of interacting defects should have randomly placed energy levels with random matrix elements for transitions between levels. This would have a number of experimental consequences such as saturation of the attenuation, inelastic phonon scattering, and absence of a phonon bottleneck. Experimental observation of the saturation of the attenuation has often been cited as evidence of the existence of two level systems in glasses. However, one of my students (Mariana Guerrero) has shown that saturation of the attenuation can occur even when there are an infinite number of levels.

My research group has also been studying interacting systems of randomly placed magnetic (vector) dipoles. An early motivation of this work were some low frequency susceptibility measurements which showed striking changes in behavior as the concetration of dipoles was varied.[1] We can qualitatively explain the experiments with the following picture. As magnetic spins are added to the system, they form strongly interacting clusters. At low concentrations there will be a broad distribution of barrier heights associated with small clusters of strongly interacting spins in a variety of environments. At higher concentrations there will be larger clusters, each having a greater variety of spin configurations and barrier heights than is available to the smaller clusters. Since the lowest barrier will dominate the dynamics of a cluster, higher concentrations will have a narrower effective distribution of barrier heights. This work has been published in Solid State Communications[2] and a reprint is enclosed.

Little is know about dipolar spin glasses even though they are important experimentally. The previous paragraph is just one example. Interacting vector dipoles also provides an interesting contrast to my tensor dipole calculation. Because of this, my student (Wen Wang) and my postdoc (Eric Grannan) are currently working on Monte Carlo calculations of dipolar spin glasses.

To summarize, my research group has been studying defects interacting via long range magnetic and elastic interactions in an effort to better understand glasses and disordered systems.




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# POSSIBLE EXPLANATION OF SUSCEPTIBILITY EXPERIMENTS ON DILUTE SYSTEMS OF RANDOM MAGNETIC DIPOLES

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Qualitative fits to low frequency susceptibility measurements of a random system of magnetic dipoles is consistent with the following picture. As we increase the concentration, larger spin clusters will appear which will have available a greater variety of spin configurations and barrier heights. Since the lowest barrier height dominates the dynamics of a cluster, the higher concentration samples have lower effective barrier heights.

The interplay of randomness and interactions is responsible for a wealth of physical phenomena found in disordered systems. Spin glasses and random magnets are well known examples. Materials in which one can vary the concentration of dipoles can afford great insight into the nature of these systems. One such material is $LiHo_xY_{1-x}F_4$ which is a crystalline insulator with a $CaWO_4$ structure[1] whose rare earth sites are randomly occupied by either magnetic $Ho^{3+}$ or non-magnetic $Y^{3+}$ ions. Due to crystal-field splitting, the ground state of an isolated $Ho^{3+}$ is a time-reversed non-Kramers doublet that can be thought of as an Ising spin.[1-3] ("Non-Kramers" means that $Ho^{3+}$ has an even number of f-electrons.) The first excited crystal field level lies 9.4 K above the doublet. Because the Ising-like magnetic moments are large ($\mu_{eff} = 7\mu_B$) and far apart, the dipolar interactions dominate even the nearest neighbor superexchange interactions.[1] For the diagonal part of the Hamiltonian, the exchange energy for nearest neighbor $Ho^{3+}$ ions is half that of the dipolar energy, and for next nearest neighbors it is only 5% of the dipolar interaction. The magnitude of the interactions is reflected in the fact that when x=1, a ferromagnetic transition occurs at $T_c$=1.53 K. [1]

Recent experiments[4-6] on the frequency dependent magnetic susceptibility have shown dramatic changes in behavior as the concentration of $Ho^{3+}$ was varied. Most striking are the normalized plots, shown in Figure 1, of $\chi''/\chi''_p$ versus $\log(f/f_p)$ where $\chi''$ is the imaginary part of the susceptibility, $\chi''_p$ is the maximum value of $\chi''$, and $f_p$ is the frequency at which the peak occurs. The frequency f was low and ranged between 0.2 Hz and 20 kHz. For x=0.167 the curves *broadened* symmetrically as the temperature was lowered from 250 mK to 150 mK. Broadening with decreasing temperature, though not necessarily symmetric, is often seen in both structural glasses and spin glasses above their freezing temperature $T_g$ when the appropriate response functions are measured. Such behavior is associated with slow relaxation processes described in terms of stretched exponentials which have the form $\exp(-(t/\tau)^\beta)$.[7] Fits to the data often require $\beta$ to decrease slightly with decreasing temperature. In frequency space, this corresponds to broadening of $\chi''(\omega)/\chi''_p$ curves. In contrast with this, for x=0.045, the curves *narrowed* as the temperature decreased from 300 mK to 150 mK. This behavior was so bizarre that initial fits to the data with temperature dependent parameters led to the interpretation that the number of high barriers to spin relaxation was *decreasing* as the temperature decreased.[5,6] However, as we shall show, such behavior is consistent with an effective decrease in the barrier height with increasing concentration.

To understand the data, we should keep in mind that the susceptibility measurements are made by applying an alternating magnetic field along the z-axis and watching how the sample magnetization follows. Since the total magnetization is not conserved, phonons, rather than spin-conserving interactions, are most likely responsible for spin-flip processes. Thus the dynamics involves not only random interactions between spins, but also the coupling between spins and phonons. The fact that the peak frequencies of $\chi''$ obey an Arrhenius law at both concentrations implies that the spin-lattice coupling results in dynamics that is activated.

We propose that the dynamics is determined by clusters of strongly interacting spins. Each cluster has a characteristic frequency $\omega_p = \omega_o \exp(-\Delta/kT)$, where $\Delta$ is the barrier height that the cluster must surmount in changing configurations. At low concentrations, there are predominantly small clusters in a variety of environments and, as a result, there is a broad

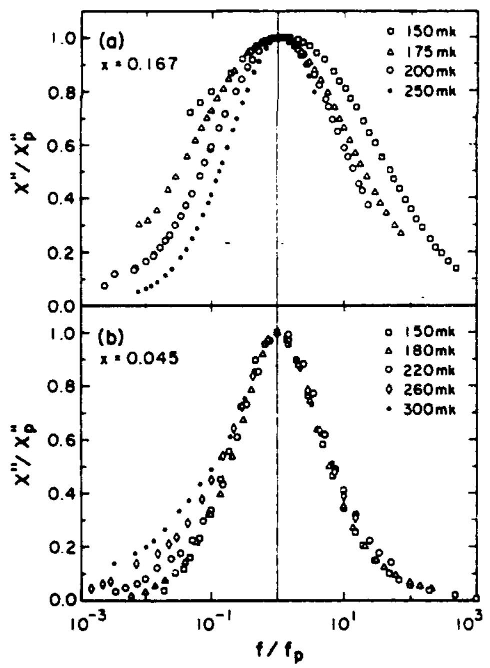


Fig. 1: Experimental data from Refs. 4 and 5 of the imaginary part of the magnetic susceptibility normalized by peak frequency and amplitude for $LiHo_xY_{1-x}F_4$ with x=0.167 and x=0.045. $\chi''$ broadens (narrows) with decreasing temperature for the more concentrated (dilute) sample.

distribution of barrier heights. At higher concentrations the spin clusters are larger. These larger clusters have more configurations available to them and hence, a greater variety of barriers from which to choose. The dynamics of a cluster will be dominated by the lowest barrier available. Thus the higher concentration sample will have effectively smaller barrier heights. If we talk in terms of a distribution of barrier heights, then we expect the higher concentration sample to have a narrower effective distribution on the high barrier side.

The evidence that there are many small clusters at low concentrations comes from the specific heat for x=0.045 where there is a Schottky peak at 0.3 K.[6] The peak accounts for about 20% of the electronic entropy. If the peak is due to a collection of degenerate, noninteracting two level systems, a peak at 0.3 K implies an energy splitting of about 0.7 K, which is comparable to the nearest neighbor coupling deduced from specific heat measurements on $LiHoF_4$.[1] The magnitude of the nearest neighbor dipolar coupling is 0.61 K and that of superexchange is −0.34 K. Thus the Schottky peak probably comes from nearest neighbor interactions splitting the ground state doublets of two or three $Ho^{3+}$ ions. It is probable that contributions to the electronic entropy outside of the Schottky peak come from small clusters in different environments.

Evidence that higher concentrations have a large number of configurations can be seen in the broad bump in the specific heat for x=0.167. Such a bump is characteristic of spin glasses.

We now show that the $\chi''(\omega)$ susceptibility data agrees with these expectations. We assume that both the x=0.045 and x=0.167 systems consist of spin clusters with collective modes, each of which has a resonant frequency $\omega_p$ and a relaxation rate reflected in a linewidth $\Gamma$. Since the spin dynamics seen in the susceptibility measurements is due to phonons, the relaxation rate should also be due to spin-phonon processes. In small clusters we suspect that the relaxation is due to the Orbach process[8] in which a transition between levels A and B of a ground state doublet occurs via a resonant excitation to a higher state C. Suppose $\Delta_\Gamma$ is the energy difference between levels B and C and that the system is in state B. Then in the Orbach process, a phonon of energy $\Delta_\Gamma$ is absorbed and the system makes a transition to C. This is followed by a de-excitation to A with the emission of a phonon whose energy equals the energy difference between C and A. This process results in an exponential temperature dependence $\Gamma = \Gamma_o \exp(-\Delta_\Gamma/kT)$ and is known to dominate single ion spin relaxation in dilute samples of paramagnetic ions at low temperatures. In small clusters the ground state doublets combine to form levels whose typical spacing is on the order of the nearest neighbor coupling. We can use the Schottky peak seen in the specific heat at T=0.3 K for x=0.045 to deduce a value for $\Delta_\Gamma$. Since $\Delta_\Gamma$ will be somewhat larger than the temperature of the peak, we will take $\Delta_\Gamma \sim 0.6$ K. In larger clusters found primarily at higher concentrations, phonon modulation of the barrier height may be a source of relaxation. However, to avoid using a different functional form for different concentrations, we will use $\Gamma = \Gamma_o \exp(-\Delta_\Gamma/kT)$ for all concentrations.

The simplest form for the susceptibility $\chi_1''(\omega)$ of a cluster comes from the Debye model of relaxation which follows from the Bloch equations. Unfortunately the traditional Debye model cannot fit the data because it predicts that $\chi_1''(\omega)/\chi_p''$ versus $\omega/\omega_p$ is independent of temperature.[5,6] Thus the simple Bloch equations cannot be used to describe the dynamics of the magnetization. The source of the problem is the fact that the Debye susceptibility has a peak at $\omega\tau = 1$, i.e., the peak and the width of the Lorentzian are coupled together. Describing the data requires having the peak and the width independent of each other. We therefore phenomenologically model the susceptibility $\chi_1''(\omega)$ of a cluster with a simple Lorentzian form which has this feature:

$$\chi_1''(\omega) \propto \left\{ \frac{\Gamma}{(\omega-\omega_p)^2+\Gamma^2} - \frac{\Gamma}{(\omega+\omega_p)^2+\Gamma^2} \right\} \quad (1)$$

where $\omega_p = \omega_o \exp(-\Delta/kT)$. $\chi_1''(\omega)$ is odd in $\omega$ and together with $\chi_1'(\omega)$ obeys the Kramers-Kronig relations. We investigated frequency and temperature ranges covering those examined experimentally. We varied $\omega_o$ and $\Gamma_o$ from $\sim 1$ Hz to $10^{11}$ Hz, and the temperature T from 100 to 500 mK.

The response of the system as a whole is given by averaging $\chi_1''(\omega)$ over an appropriate distribution function. We have chosen to examine two different distribution functions. One is the probability distribution of barrier heights $\Delta$ and the other is the distribution of the prefactor $\omega_o$. This leads to

$$\chi''(\omega) = A\int_0^\infty dx P(x)\chi_1''(\omega) \qquad (2)$$

where $x$ is $\omega_o$ or $\Delta$. $P(x)$ is a gaussian distribution given by

$$P(x) = \frac{1}{\sqrt{2\pi}\sigma_x}\exp\left[-\frac{(x-\bar{x})^2}{2\sigma_x^2}\right] \qquad (3)$$

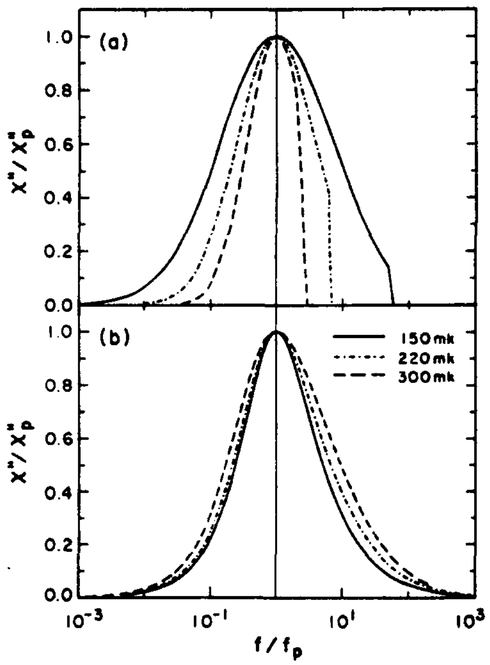


**Fig. 2:** Theoretical curves of the imaginary part of the magnetic susceptibility normalized by peak frequency and amplitude. These are calculated by averaging the individual spin modes over a distribution of barrier heights. The different temperature dependence in (a) and (b) is the result of changing only one parameter, the width of the distribution $\sigma_\Delta$: (a) $\sigma_\Delta$=0.3 K and (b) $\sigma_\Delta$=0.8 K. The rest of the input parameters are the same: $\Gamma_o = 10^3$ Hz, $\omega_o = 10^5$ Hz, $\bar{\Delta} = 0$ K, and $\Delta_\Gamma = 0.6$ K. (The steep drop at high frequencies is an artifact of the absence of barriers below the peak in the distribution P($\Delta$). If we shift the peak position ($\bar{\Delta} > 0$) and again integrate over positive barrier heights, then the steep drop disappears.)

$\bar{x}$ is the mean value of $x$. Looking at different distributions helps us to avoid attaching too much physical significance to a given parameter and allows us to look for general trends. For each distribution we allow *only one* parameter to vary as the concentration changes. For a given concentration all the input parameters were the same at all temperatures. There are no temperature dependent input parameters and the distribution $P(x)$ has no temperature dependence. Nonetheless, depending on the concentration, the normalized plots of $\chi''(\omega)$ have the same qualitative features as the experiments, i.e., they either broaden or narrow with decreasing temperature.

We now examine the relevant parameter which can be tuned in such a way as to have the same effect as changing the concentration. In the case of averaging over a distribution of barrier heights, the relevant parameter is the width $\sigma_\Delta$ of the distribution. The result of varying $\sigma_\Delta$ is shown in Figure 2. As expected, a broad distribution corresponds to low concentrations where $\chi''(\omega)/\chi''_p$ narrows as T decreases, while a narrow distribution corresponds to higher concentrations and produces susceptibility curves which broaden with decreasing temperature. Since our arguments for a narrower width pertain only to the barrier height distribution above the peak $\bar{\Delta}$, we set $\bar{\Delta} = 0$ and integrate only over positive barrier heights.

Averaging over the attempt frequency $\omega_o$ corresponds to averaging over the curvature of the different potential wells. In this case the barrier height $\Delta$ measured relative to $\Delta_\Gamma$ is the relevant parameter. As the example in Figure 3 shows, high barriers ($\Delta > \Delta_\Gamma$) correspond to low concentrations while low barriers correspond to high concentrations ($\Delta < \Delta_\Gamma$). This agrees with our previous arguments. The advantage of integrating over a distribution of prefactors $\omega_o$ is that the peak frequencies $f_p$ of $\chi''(\omega)$ obey an Arrhenius law (as is seen experimentally[5,6]), whereas averaging over barrier heights $\Delta$ leads to temperature dependent deviations from Arrhenius behavior. This can be easily seen by replacing $\chi_1''(\omega)$ with infinitely narrow delta functions $\chi_D''(\omega) \propto [\delta(\omega-\omega_p) - \delta(\omega+\omega_p)]$ and integrating over the appropriate distribution. Since $\chi_1''$ is much narrower than either P($\omega_o$) or P($\Delta$), the delta function approximation should be a good one. If we integrate over prefactors, $f_p = \bar{\omega_o}\exp(-\Delta/T)$, whereas integrating over barrier heights leads to $f_p = \omega_o \exp(-(\bar{\Delta}/T) - (\sigma_\Delta/T)^2)$.

We see from Figures 1 to 3 that for x=0.167, both the experimental and theoretical curves broaden symmetrically as T decreases. For x=0.045, however, the experimental $\chi''/\chi''_p$ plots narrow at frequencies below the peak but not above. The theoretical curves narrow in a more symmetric fashion. It is not clear that this discrepancy has any fundamental physical significance since the precise form of the narrowing depends to some extent on the choice of a demagnetization correction that is needed to

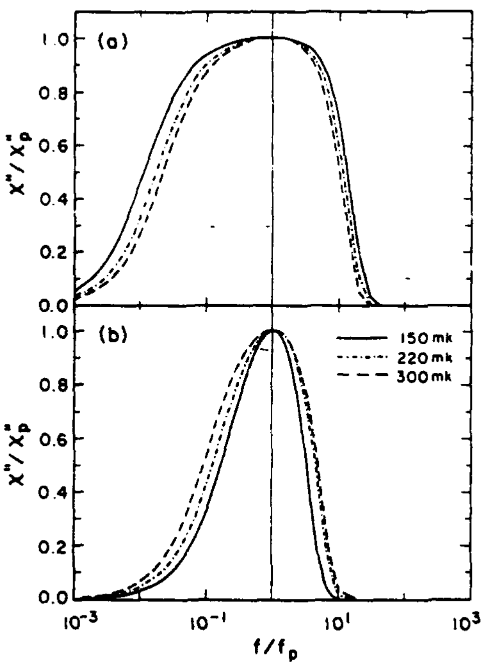


Fig. 3: Theoretical curves of the imaginary part of the magnetic susceptibility normalized by peak frequency and amplitude. These are calculated by averaging the individual spin modes over a distribution of Arrhenius prefactors. The different temperature dependence in (a) and (b) is the result of changing only one parameter, the activation energy associated with the single cluster peak frequency $\Delta$: (a) $\Delta$=0.3 K and (b) $\Delta$=1.0 K. The rest of the input parameters are the same: $\Gamma_o = 10^5$ Hz, $\overline{\omega_o} = 10^5$ Hz, $\Delta_\Gamma = 0.6$ K, and $\sigma_{\omega_o} = 10^{-4}$ K.

compensate for the shape of the sample.[9] A quantitative fit to the data would no doubt require averaging over a distribution function more complicated than a gaussian.

We should point out that a finite value of the single cluster linewidth $\Gamma$ is required to obtain the proper temperature dependence of the theoretical curves. What happens if $\Gamma$=0 can be seen by again replacing $\chi''_1$ with the infinitely narrow delta functions in $\chi''_D$. If we average over barrier heights, $\chi''/\chi''_p$ broadens with decreasing temperature when plotted versus $\log(f/f_p)$. On the other hand, if we integrate over a distribution of prefactors, $\chi''/\chi''_p$ versus $(f/f_p)$ is temperature independent. Thus without $\Gamma$ one can no longer model the change in behavior with concentration. One can in fact associate this behavior with a single cluster relaxation rate which decreases as the concentration increases. However, the physics behind this viewpoint is not entirely clear.

To summarize, we have performed qualitative fits of ac susceptibility measurements on samples of random magnetic dipoles. We find that tuning a single temperature independent parameter has the same effect as is seen experimentally when the concentration is varied. Our results are consistent with the following picture. The dynamics is activated and involves random interactions between spins as well as interactions between spins and phonons. At low concentrations there will be a broad distribution of barrier heights associated with small clusters of strongly interacting spins in a variety of environments. At higher concentrations there will be larger clusters, each with a variety of available spin configurations and barrier heights. Since the lowest barrier will dominate the dynamics of a cluster, higher concentrations will have a narrower effective distribution of barrier heights.

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